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Atmospheric optical and radiative effects of anthropogenic aerosol constituents from India

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Abstract

A box model has been used to compare the burdens, optical depths and direct radiative forcing from anthropogenic $\text{PM}_{2.5}$ aerosol constituents over the Indian subcontinent. A $\text{PM}_{2.5}$ emission inventory from India for 1990, compiled for the first time, placed anthropogenic aerosol emissions at 12.6 Tg yr^{-1} . The contribution from various aerosol constituents was 28% sulphate, 25% mineral (clay), 23% fly-ash, 20% organic matter and 4% black carbon. Fossil fuel combustion and biomass burning accounted for 68% and 32%, respectively, of the combustion aerosol emissions. The monthly mean aerosol burdens ranged from 4.9 to 54.4 mg m^{-2} with an annual average of $18.4 \pm 22.1 \text{ mg m}^{-2}$. The largest contribution was from fly-ash from burning of coal (40%), which has a high average ash content of 30%. This was followed by contributions of organic matter (23%) and sulphate (22%). Alkaline constituents of fly-ash could neutralise rainfall acidity and contribute to the observed high rainfall alkalinity in this region. The estimated annual average optical depth was 0.08 ± 0.06 , with sulphate accounting for 36%, organic matter for 32% and black carbon for 13%, in general agreement with those of Satheesh et al. (1999). The mineral aerosol contribution (5%) was lower than that from the previous study because of wet deposition from high rainfall in the months of high emissions and the complete mixing assumption in the box model. The annual average radiative forcing was $-1.73 \pm 1.93 \text{ W m}^{-2}$ with contributions of 49% from sulphate aerosols, followed by organic matter (26%), black carbon (11%) and fly-ash (11%). These results indicate the importance of organic matter and fly-ash to atmospheric optical and radiative effects. The uncertainties in estimated parameters range 80–120% and result largely from uncertainties in emission and wet deposition rates. Therefore, improvement is required in the emissions estimates and scavenging ratios, to increase confidence in these predictions. © 2000 Elsevier Science Ltd. All rights reserved.

Keywords: $\text{PM}_{2.5}$ emission inventory; Black carbon; Organic matter; Minerals; Fly-ash; Sulphate; Box model

1. Introduction

Anthropogenic aerosol emissions to the troposphere have increased in recent decades (Houghton et al., 1996) and their effects include scattering and absorption of incident solar radiation (direct effect) and modifying the microphysical and optical properties of clouds (indirect effect). Much of the work on direct radiative forcing of aerosols has focused on sulphate aerosols (Charlson et

al., 1991, 1992; Kiehl and Briegleb, 1993; Boucher et al., 1998). However, recent global models have identified the importance of carbonaceous aerosols (Penner, 1995; Penner et al., 1992; Haywood and Ramaswamy, 1998), and mineral aerosols (Tegen et al., 1996; Sokolik and Toon, 1996). The resultant forcing depends on the relative source strengths and burdens, which have a strong regional dependence (Tegen et al., 1997). In order to refine global scale predictions, it is of importance to study aerosol emissions and atmospheric effects on a regional scale.

In previous regional studies of anthropogenic sulphate (Venkataraman et al., 1999) and carbonaceous (Reddy and Venkataraman, 1999) aerosols over India, an

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aggregate emissions inventory for these species for 1990 was developed. A box model was used, along with regional meteorological parameters, to simulate atmospheric processes including reaction, deposition and advection to estimate aerosol burdens and radiative effects. Sulphate and organic matter aerosols contributed in similar magnitude to decrease in radiation flux ($-1.1 \pm 1.0 \text{ W m}^{-2}$ from sulphate and $-0.69 \pm 0.55 \text{ W m}^{-2}$ from organic matter), while black carbon aerosols caused a $+0.27 \pm 0.17 \text{ W m}^{-2}$ increase in radiation flux.

The objectives in this study were three-fold: (i) to develop a $\text{PM}_{2.5}$ emissions inventory for India of particles smaller than $2.5 \mu\text{m}$ diameter, (ii) to estimate burdens and radiative effects of anthropogenic fly-ash and mineral aerosols, and (iii) to compare the relative contribution of sulphate, fly-ash, mineral and carbonaceous aerosols to optical depth and radiative forcing.

2. Modelling approach

2.1. Model description

Estimates of aerosol burden (of sulphate, carbonaceous, fly-ash and mineral aerosols), optical depth and radiative forcing over India are made using regional emissions and atmospheric parameters. A carbonaceous aerosol and fly-ash emission inventory is compiled from aggregate fossil fuel (industrial coal and petroleum) combustion and biomass burning (previously used for a SO_2 emissions inventory) along with appropriate emission factors. An estimate of anthropogenic mineral aerosol emissions from disturbed soils is made from reported mineral emission fluxes over India (Tegen and Fung, 1995). Aerosol lifetimes and burdens are calculated using a box model considering aerosol transport and removal mechanisms including advection, wet deposition and dry deposition. This follows from the previous study of formation and direct radiative forcing from sulphate aerosols over the Indian subcontinent (Venkataraman et al., 1999). The box model used covers an area ($L \times W$) of $7.3 \times 10^{12} \text{ m}^2$ ($8\text{--}36^\circ\text{N}$ and $70\text{--}92^\circ\text{E}$), including the land-mass of India and surrounding oceans, with an average mixing height of $0.5\text{--}2.0 \text{ km}$ (Charlson et al., 1992) and with uniformly mixed constituents. The Fourier amplitude sensitivity test (FAST) (Cukier et al., 1973; McRae et al., 1982) is used to average monthly mean meteorological data from 18 stations spread uniformly over India to obtain spatially averaged aerosol lifetime, burden, optical depth and radiative forcing. The meteorological parameters are normally distributed about their monthly means with an additive search curve for small monthly range or uncertainty and an exponential search curve for a range exceeding an order of magnitude.

2.2. Emissions

2.2.1. Fossil fuel combustion

The fossil fuel-use data for 1990 were compiled from Mitra (1992), also previously used for sulphur emission estimates (Venkataraman et al., 1999) (Table 1). Black carbon (BC) emission factors are from Bocola and Cirillo (1989) and USEPA (1996a), used previously by Cooke and Wilson (1996) for global black carbon emission inventories. Primary organic carbon (OC) aerosols are emitted from the same combustion sources as black carbon, while secondary organic carbon is formed from atmospheric reactions of gaseous volatile organic compounds (VOCs) (Turpin and Huntzicker, 1991) and is predominantly sub-micron in size (McMurry and Zhang, 1989). Organic carbon emission factors are developed from black carbon emission factors multiplied by an appropriate OC/BC ratio, following previous global organic matter emission estimates (Liousse et al., 1996). We use a previously suggested BC/OC ratio of 0.30, based on the measurements of black carbon and organic carbon concentrations in aerosols collected near source regions (Pierson and Russel, 1979; Rosen et al., 1980; Cadle and Dasch, 1988; Brémond et al., 1989). From the measurements of Countess et al. (1981), the ratio of organic matter (OM) to organic carbon is assumed 1.3, to account for associated hydrogen, oxygen, and minor amounts of other species. From the above assumptions, the ratio of aerosol organic matter to black carbon (OM/BC) is 4.33.

The black carbon and organic matter emission factors developed above correspond to particles smaller than $2 \mu\text{m}$ diameter. Fly-ash emission factors, in particles smaller than $2.5 \mu\text{m}$ diameter, are obtained by subtracting the sum of BC and OM from $\text{PM}_{2.5}$ emission factors for all the fossil fuels. This follows from our assumption that $\text{PM}_{2.5}$ emissions from fossil-fuel combustion consist of black carbon, organic matter and fly-ash. Basu and Chakrabarti (1990) report the ash content of coal used in different Indian industrial sectors and this was used to generate $\text{PM}_{2.5}$ emission factors from the USEPA (1996a) compilation as a function of coal ash-content. For liquid and gaseous fossil fuels, $\text{PM}_{2.5}$ emission factors were given as mass of pollutant per mass of fuel burnt (USEPA, 1996a; Bocola and Cirillo, 1989).

2.2.2. Biomass burning

Biomass burning in India includes wood, agricultural residue and dung-cakes used as fuel in cooking stoves as well as forest and grassland firing (Joshi, 1991). 1989–1990 estimates of biomass burning (Kaul, 1993; Kaul and Shah, 1993; Sinha and Joshi, 1997) are used for carbonaceous aerosols and fly-ash emission estimates. Measured total particulate matter (PM) emission factors for fuelwood, agricultural residue and dung-cakes burned in cooking stoves are from Joshi et al. (1989, 1991).

Table 1
Fuel use data and emission estimates of carbonaceous aerosols and fly-ash for 1990

Source	Consumption	Mean emission factors (g kg ⁻¹)			Aerosol emissions (Tg yr ⁻¹)			
	(MT yr ⁻¹)	PM _{2.5} ^a	BC ^a	OM ^a	PM _{2.5}	BC	OM	Fly-ash
Fossil fuels								
<i>Coal</i>								
Steel	22.3 ^b	8.840 ^c	1.000 ^d	4.333 ^e	0.197	0.022	0.097	0.078
Power	135.7	17.160	1.000	4.333	2.329	0.136	0.588	1.605
Railways	6.5	10.400	1.000	4.333	0.068	0.007	0.028	0.033
Cement	11.5	13.000	1.000	4.333	0.150	0.012	0.050	0.088
Sponge iron	1.0	13.000	1.000	4.333	0.013	0.001	0.004	0.008
Fertiliser	5.5	10.400	1.000	4.333	0.057	0.006	0.024	0.028
Coke	3.5	20.800	1.000	4.333	0.073	0.004	0.015	0.054
Others	31.7	15.600	1.000	4.333	0.495	0.032	0.137	0.325
Lignite	11.0	5.860	1.000	4.333	0.064	0.011	0.048	0.006
<i>Petroleum</i>								
LPG	2.3	0.060	0.060	0.260	0.000	0.000	0.001	-
Kerosene	8.2	0.300	0.300	1.300	0.002	0.002	0.011	-
H S Diesel	20.7	3.050	0.700	3.033	0.063	0.014	0.063	-
Fuel oils	8.8	2.776	0.100	0.433	0.024	0.001	0.004	0.020
Motor Gas.	3.5	1.800	0.000	0.000	0.006	0.000	0.000	0.006
Aviation gas	1.8	1.800	0.000	0.000	0.003	0.000	0.000	0.003
Sub-total					3.345	0.247	1.069	2.255
						± 0.229 ^f	± 0.990 ^f	± 1.336 ^g
Biomass								
Wood	252.1 ^h	2.112 ⁱ	0.285 ^j	1.290 ^k	0.532	0.072	0.325	0.135
Dung-cakes	106.1	6.512	0.326	3.979	0.691	0.035	0.422	0.234
Agr. Res.	99.2	4.048	0.457	2.473	0.402	0.045	0.245	0.111
Forest	26.7	18.000 ^l	1.530	12.402	0.481	0.041	0.331	0.109
Grassland	5.2	18.000	1.530	12.402	0.094	0.008	0.064	0.021
Sub-total	489.3				2.199	0.201	1.388	0.610
						± 0.117 ^m	± 0.777 ^m	± 0.367 ^m
Total					5.74	0.45 ± 0.26	2.46 ± 1.26	2.86 ± 1.39

^aPM_{2.5}: emissions of particulate mass smaller than 2.5 µm diameter, BC: black carbon, OM: organic matter.

^bFossil fuel use data (Mitra, 1992).

^cPM_{2.5} emission factors for fossil fuels (USEPA, 1996a); coal ash content (Basu and Chakrabarti, 1990).

^dBC emission factors for fossil fuels (Bocola and Cirillo, 1989; USEPA, 1996a).

^eOM emission factors for fossil fuels average BC/OC = 0.3 (Pierson and Russel, 1979; Rosen et al., 1980; Cadle and Dasch, 1988; Brémond et al., 1989); OM/OC = 1.3 (Countess et al., 1981).

^fAssumed uncertainty in emission factors of BC and OM from coal and lignite are 100%.

^gAssumed uncertainty in emission factors of PM_{2.5} from coal and lignite are 60% and hence uncertainty in emission factors of fly-ash are 60%.

^hBiomass use data (Kaul, 1993; Kaul and Shah, 1993; Sinha and Joshi, 1997).

ⁱPM_{2.5} emission factors for biomass fuels: PM emission factors (Joshi, 1989, 1991); average PM_{2.5}/PM = 0.80 (Dasch, 1982; Jenkins et al., 1993; Radke et al., 1988, 1991).

^jBC emission factors for biofuels, forest and grassland burning: BC/PM_{2.5} (Lioussé et al., 1996).

^kOC/PM_{2.5} = 0.45 (Patterson and McMahon, 1984; Dasch, 1982; Cooper, 1980); OM/OC = 1.3 (Countess et al., 1981).

^lPM_{2.5} emission factors for forest and grassland burning (Ward et al., 1991; Patterson and McMahon, 1984; Patterson et al., 1986).

^mAssumed uncertainty in emission factors of PM from biofuels are 75% and hence uncertainty in emission factors of BC, OM and fly-ash are 75%. Assumed uncertainty in emission factors of PM_{2.5} from forest firing and grassland burning are 56% and uncertainty in emission factors of BC, OM and fly-ash are 56%.

PM_{2.5} emission factors are calculated from these using a PM_{2.5}/PM ratio of 0.80 reported from wood and agricultural residue burning experiments (Dasch, 1982;

Jenkins et al., 1993; Radke et al., 1991). PM_{2.5} emission factors for forest and grassland firing are taken from Ward et al. (1991), Patterson and McMahon (1984) and

Patterson et al. (1986). Emission factors for black carbon are calculated from BC/PM_{2.5} ratios compiled by Lioussé et al. (1996) for different biomass types (Table 1). OC/PM_{2.5} ratios are taken for burning of fuel wood, agricultural residue, forest and grasslands as 45% (Patterson and McMahon, 1984; Dasch, 1982; Cooper, 1980) and assumed the same for burning of dung-cakes. Once again, the ratio of organic matter to organic carbon is assumed 1.3 (Lioussé et al., 1996). Fly-ash emissions from biomass burning are estimated as for fossil fuels, by deducting the sum of black carbon and organic matter fraction from the PM_{2.5} emissions. While, emissions from the biomass burning vary seasonally, as a first estimate, it is assumed that emissions are uniformly distributed throughout the year.

2.2.3. Anthropogenic carbonaceous and fly-ash emissions

Estimated black carbon emissions from India for 1990 are 0.45 Tg yr⁻¹, with 55% from fossil fuel combustion and 45% from biomass burning. These are larger than previous estimates of black carbon emissions from India of 0.26 Tg yr⁻¹ (Cooke, 1999) for the base year 1984–1985 (these exclude agricultural fires) from increase in fuel use between 1984–1985 and 1990. For comparison, there are three previous estimates of global black carbon emission inventories for the base year 1984–1985 (Cooke and Wilson, 1996; Lioussé et al., 1996; Penner et al., 1993). These range 12.3–13.5 Tg yr⁻¹ with 2.22–5.28 Tg yr⁻¹ from Asia.

Organic matter emissions from India are estimated at 2.46 Tg yr⁻¹ for 1990, with fossil fuel combustion and biomass burning contributing 43 and 57%, respectively (Table 1). In comparison, estimates of global organic matter emissions are 73.20 Tg yr⁻¹ from the above sources for 1984–1985, with fossil fuel and biomass contributing 39 and 61%, respectively (Lioussé et al., 1996). From the Cooke and Wilson (1996) estimates of BC discussed earlier, using the appropriate OM/BC ratios the global organic matter emissions would be 80.70 Tg yr⁻¹ with Asia contributing 10.40 Tg yr⁻¹, in agreement with that of Lioussé et al. (1996).

Fly-ash emissions from India are 2.86 Tg yr⁻¹, with fossil fuels and biomass burning accounting for 79 and 21%, respectively. The major contribution to fly-ash is from coal combustion (2.23 Tg yr⁻¹), with the power sector alone contributing 1.61 Tg yr⁻¹. This is because of the high ash content of coal (33%) used in this sector (Basu and Chakrabarti, 1990) which accounts for 59% of total coal consumption (Mitra, 1992).

2.2.4. Anthropogenic mineral aerosols

A number of global mineral aerosol emission estimates have been made from both natural (wind-blown dust) and anthropogenic (land disturbance) origin, with more recent estimates significantly larger than earlier estimates

Table 2

Mineral aerosols emission estimates

Parameter	Value
Emission flux ^a (kg m ⁻² yr ⁻¹)	0.00–0.05
% of continental area as a source	4.50
Source area (m ²)	2.46 × 10 ¹¹
Total emissions (1–10 µm) (Tg yr ⁻¹)	6.16
PM _{2.5} emissions ^b (Tg yr ⁻¹)	3.142 (0.0–6.16) ^c

^aTegen and Fung (1995).

^bPM_{2.5}/total emissions = 0.51 (Tegen and Lacis, 1996).

^cValue in the parenthesis is range of mineral aerosol emissions.

(d'Almeida, 1986; Tegen and Fung, 1994, 1995; Duce, 1995). We have developed an estimate for mineral aerosols of size fraction less than 2.5 µm (PM_{2.5}) from anthropogenic activities. This is based on the reported mineral aerosol fluxes (Table 2) over the Indian region multiplied by appropriate land area which includes cultivated eroded soils, uncultivated eroded soils, recently cultivated areas and recently deforested areas (Tegen and Fung, 1995). The reported mineral fluxes over India are 0–0.05 kg m⁻² yr⁻¹ and the area affected by anthropogenic disturbance is estimated at 1.11 × 10¹⁰ m². This gives annual mineral aerosol emissions of 6.16 Tg yr⁻¹, distributed 51 and 49% between the 0–2.5 and 2.5–10 µm size fractions (Tegen and Lacis, 1996). The seasonal variation in mineral aerosol emissions over India is taken from GCM estimates of Tegen (1999). Anthropogenic mineral aerosol emissions were highest in June and July (~ 56%) with an additional 23% in April and May. This is expected because agricultural activities coincide with the dry season during this period in India.

Total anthropogenic combustion PM_{2.5} emissions (diameter range of 0–2.5 µm) of carbonaceous, fly-ash and sulphate (calculated assuming sulphur dioxide to sulphate conversion ratio of 0.61 from Venkataraman et al., 1999) from India for 1990 are 9.4 Tg yr⁻¹, assuming no emission control. Including the PM_{2.5} mineral aerosol emissions, this gives a total PM_{2.5} emissions of 12.6 Tg yr⁻¹ for India for 1990 (Fig. 1) with the largest contribution from sulphate aerosols (28%), mineral aerosols (25%) and fly-ash (23%). We compared our estimate with a previous PM₁₀ emission estimate (Hidy and Wolf, 1995) from a similar combustion source enumeration for India. They assumed a 10% ash content of coal (against our average of about 30%) and a 10% emission control (against our assumption of no emission control) and estimated PM₁₀ of 12.5 Tg yr⁻¹ for 1990. Assuming a PM_{2.5}/PM₁₀ ratio of 0.58 (USEPA, 1996b, c), our estimated PM₁₀ emissions would be 16.2 Tg yr⁻¹. Our estimate is about 30% higher than that of Hidy and Wolf (1995) and is based on a realistic ash-content for coal and an assumption of no emissions control, which represents worst-case emissions.

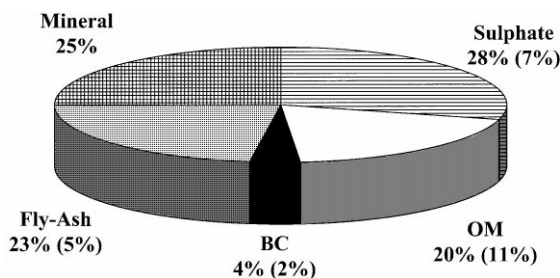


Fig. 1. Relative contribution of aerosol constituents to $PM_{2.5}$ aerosol emissions from India for 1990 (total 12.6 Tg yr^{-1}). Emissions from fossil-fuel combustion and biomass burning (in parenthesis) contribute to sulphate, organic matter (OM), black carbon (BC) and fly-ash; mineral aerosols are the clay-fraction from anthropogenic land disturbance.

2.2.5. Uncertainties in emissions

Recent measurements of the emission of sub-micron light absorbing particles from lignite power plant (Bond et al., 1998) suggest an emission factor, for sub-micron black carbon, of 0.05 g kg^{-1} , which is a factor 20 lower than that assumed here. Further, emissions uncertainties have shown to have a significant effect on the predicted single-scatter albedo (Heintzenberg et al., 1997). In order to account for these effects, we have included an uncertainty of 100% in BC emissions from fossil fuels (which includes the reduced emission factor suggested). In addition, a 75 and 55% uncertainty was assumed (Joshi et al., 1989, 1991; Ward et al., 1991; Patterson et al., 1986; Patterson and McMahon, 1984) for BC emissions from biofuels (fuelwood, agricultural residue and dung-cakes) and forest fires, respectively. This gave a black carbon emission range of $0.45 \pm 0.26 \text{ Tg yr}^{-1}$ (Table 1). Organic matter (OM) emissions are derived from BC and have the same uncertainties resulting in OM emissions of $2.46 \pm 1.26 \text{ Tg yr}^{-1}$. Assumed uncertainties in emission factors of fly-ash from coal and lignite are 60% (USEPA, 1996a) and 75% for biofuel combustion (Joshi et al., 1989, 1991) and 56% for forest fires and grassland firing (Ward et al., 1991; Patterson et al., 1986; Patterson and McMahon, 1984). Estimated annual fly-ash emissions range $2.86 \pm 1.39 \text{ Tg yr}^{-1}$. These uncertainties in the aerosol emissions are propagated in the model calculations and a sensitivity analysis carried out to assess the influence of different input parameters on the estimated radiative forcing.

2.3. Removal mechanisms and deposition parameters

The transport and removal mechanisms considered are advection, dry deposition and wet deposition. The lifetime (L_t) of aerosols in the box is given as

$$L_t = \frac{1}{k} = \frac{1}{k_a} + \frac{1}{k_d} + \frac{1}{k_w}, \quad (1)$$

where k_a is the advection removal rate (s^{-1}), k_w is the wet deposition rate (s^{-1}) and k_d is the dry deposition rate (s^{-1}). k_a , k_w and k_d are given as

$$k_a = \frac{1}{\left(\frac{v \cos \theta}{L} + \frac{v \sin \theta}{W} \right)}, \quad (2)$$

where v is the surface wind speed (m s^{-1}), θ is the average wind direction, L is the length of the box (m) and W is the width of the box (m);

$$k_w = \frac{RS\rho_{\text{water}}}{H\rho_{\text{air}}}, \quad (3)$$

where R is the monthly mean rainfall (m s^{-1}), S is the scavenging ratio, ρ_{water} is the density of water (g cm^{-3}) and ρ_{air} is the density of air (g cm^{-3});

$$k_d = v_d/H, \quad (4)$$

where v_d is the dry deposition velocity of aerosols (m s^{-1}) and H is the mixing height (m).

Advection of aerosols out of the box is assumed to occur by monthly mean horizontal surface winds, with surface wind speed data for 1990 obtained from 18 meteorological stations spread across India, run by Indian Meteorological Department (IMD). An average dry deposition velocity of 0.1 cm s^{-1} (Table 3) is used for black carbon and organic matter aerosols, consistent with earlier carbonaceous aerosols transport models (Liousse et al., 1996; Cooke and Wilson, 1996; Penner et al., 1993). The dry deposition velocity of mineral aerosols depends upon particle size (Tegen and Fung, 1994). For the sub-micron fraction, the mean dry deposition velocity was 0.018 cm s^{-1} , corresponding to a mean effective radius of $0.73 \mu\text{m}$ (assuming size distribution of $dN/d\log r \propto r^{-3}$) and assumed here for $PM_{2.5}$ mineral aerosols.

Wet deposition is parameterised using scavenging ratios which, for black carbon and organic matter, are 18 to 650 (mean 180 ± 170) and 35 to 1900 (mean 340 ± 430), respectively (Dasch and Cadle, 1989). Scavenging ratios for organic matter are higher than black carbon from the greater solubility of organic matter in rainwater. Scavenging ratios used are 18 to 350 and 35 to 770 for black carbon and organic matter, respectively (Table 3). Scavenging ratios for clay-sized mineral aerosols varied from 500 to 1000 and for larger mineral particles were about 300 (Buat-Ménard and Duce, 1986) in the tropical Pacific Ocean. We use scavenging ratios of 500–1000 for $PM_{2.5}$ mineral aerosols following Tegen and Fung (1994, 1995) (Table 3). Fly-ash particles are the mineral component of combustion-derived aerosols, are assumed to follow the $PM_{2.5}$ mineral aerosols deposition characteristics and have the same deposition parameters (dry deposition velocity and scavenging ratios).

Aerosol burden (B), which is an area concentration integrated from the surface of the earth to the top of the

Table 3
Aerosol deposition properties

Parameter	Range of values
<i>Dry deposition velocity, v_d</i> (cm s ^{−1})	
Black carbon, organic mater	0.1
Mineral aerosols	0.018
Fly-ash	0.018
<i>Scavenging ratio, S</i>	
Black carbon	18–350
Organic matter	35–760
Mineral aerosols	500–1000
Fly-ash	500–1000

atmosphere (mixing height), is given by (Charlson et al., 1991, 1992)

$$B = \frac{QL_t}{A}, \tag{5}$$

where *Q* is the aerosol emission rate (g day^{−1}), *L_t* is the aerosol lifetime (day) and *A* is the area of the box (m²).

2.4. Optical properties and direct radiative forcing

Direct radiative forcing is estimated assuming an optically thin layer of aerosol, where multiple reflections within the aerosol layer are neglected (Charlson et al., 1991, 1992). The change in radiative forcing in a column of air integrated from the surface of the earth to top of the atmosphere (mixing height), as a result of the change in planetary albedo occurring in the cloud-free area of the atmosphere, is given by (Charlson et al., 1991, 1992)

$$\Delta F = -\frac{1}{4}F_T(1 - A_c)T_a^2\Delta R_a, \tag{6}$$

where ΔF is the change in forcing (W m^{−2}), $(1/4)F_T$ is the incident top of the atmosphere radiative flux and *T_a* is the fraction of light transmitted by the atmospheric layer above the aerosol layer (Table 4), *A_c* is the fractional areal cloud cover, and ΔR_a is the change in planetary albedo. While this assumes that planetary albedo is enhanced only in cloud-free regions, it has been recently shown (Boucher and Anderson, 1995) that direct radiative forcing in cloudy regions may be 25% as effective as in cloud-free regions. The assumption of an optically thin aerosol layer allows ΔR_a to be related to the aerosol transmissivity and reflectivity (Twomey, 1977)

$$\Delta R_a = r + \frac{t^2R_s}{1 - R_sr} - R_s, \tag{7}$$

where *R_s* is the albedo of the surface, *r* is the reflectivity [*r* = (1 − e^{−τ})ωβ] and *t* is the transmissivity [*t* = e^{−τ} + (1 − β)(1 − e^{−τ})ω] of the aerosol layer. Here,

Table 4
Constants used in the direct radiative forcing calculations

Symbol	Parameter	Value
<i>F_t</i> (W m ^{−2})	Top of atmosphere radiative flux	1370
<i>T_a</i>	Fraction of light transmitted by the atmospheric layer above the aerosol layer	0.76
<i>R_s</i>	Fractional albedo of underlying surface	0.15
β	Backscatter fraction	0.29

β is the globally averaged fraction of the upscattered radiation (Table 4), τ is the optical depth and ω is the single-scattering albedo (ratio of mass scattering coefficient to mass extinction coefficient):

Optical depth is a product of the aerosol burden (*B*), and the mass extinction coefficient (α_e = α_s + α_a) relating to the mass scattering coefficient (α_s) and mass absorption (α_a) coefficient of the aerosols (Charlson et al., 1992):

$$\tau = \alpha B, \tag{8}$$

Black carbon absorbs the incoming solar radiation, whereas organic matter scatters it. Results from the several measurements and theoretical estimates using Mie theory have shown that the mass absorption coefficient of black carbon ranges from 6 to 10 m² g^{−1} at visible wavelengths (Rossler and Faxvog, 1979; Jennings and Pinnick, 1980). Black carbon aerosols may also scatter radiation and the measured mass scattering coefficients vary from 1.5 to 3.0 m² g^{−1} (Sloane et al., 1981). Here we use black carbon mass absorption and scattering coefficients of 7–9 m² g^{−1} and 1.75–2.0 (Table 5), respectively, consistent with the previous studies (Liousse et al., 1996; Penner et al., 1992, 1995; Tegen et al., 1997). Organic matter contributes only to scattering and measured mass scattering coefficients range from 2.5 to 5.0 m² g^{−1} (Sloane, 1991; Tangren, 1982; Patterson and McMahon, 1984). We use an organic matter mass scattering coefficient of 3–5 m² g^{−1} (Table 5) for low relative humidity conditions, consistent with the previous studies (Liousse et al., 1996; Penner, 1995; Tegen et al., 1997).

The mass extinction efficiency of black carbon aerosols is independent of relative humidity, whereas that of organic matter increases with increasing relative humidity because of its hydrophilic nature (Ducret and Cachier, 1992; Novakov and Penner, 1993). Heintzenberg et al. (1997) point out the importance of accounting for relative humidity effects on aerosol single-scatter albedo. We use an enhancement factor of 1.7 for increasing relative humidity from 30 to 85% as suggested by Kiehl and Briegleb (1993) for sulphate aerosols and used previously for

organic matter aerosols (Liousse et al., 1996; Penner, 1995).

Mineral aerosols both absorb and scatter radiation depending upon size, with smaller particles scattering more efficiently than larger ones. Tegen et al. (1997) derived the mass extinction coefficients for sub-micron mineral aerosols to be $1.0\text{--}2.0\text{ m}^2\text{ g}^{-1}$, from previous measurements of sub-micron particles near source regions (Gomes et al., 1990; d'Almeida and Schütz, 1983; Patterson and Gillette, 1977). The single-scattering albedo (ratio of mass scattering coefficient and mass extinction coefficient) of mineral aerosols ranged from 0.91 to 0.96 for sub-micron mineral aerosols (Tegen et al., 1996). We use the sub-micron mineral aerosol optical properties for $\text{PM}_{2.5}$ mineral aerosols. Fly-ash optical properties are assumed the same as those of the mineral aerosols.

3. Results and discussion

3.1. Sensitivity analysis

A sensitivity analysis was carried out using the Fourier amplitude sensitivity test (FAST) of input variables on

Table 5
Aerosol optical properties

Symbol	Parameter	Value
α_s ($\text{m}^2\text{ g}^{-1}$)	Mass scattering coefficient	
	Black carbon	1.75–2.00
	Organic matter	3.00–5.00
α_a ($\text{m}^2\text{ g}^{-1}$)	Mass absorption coefficient	
	Black carbon	7.00–9.00
α_e ($\text{m}^2\text{ g}^{-1}$)	Mass extinction coefficient	
	Mineral aerosols	1.0–2.0
	Fly-ash	1.0–2.0
ω ($\alpha_s/\alpha_s + \alpha_a$)	Single scattering albedo	
	Mineral aerosols	0.907–0.964
	Fly-ash	0.907–0.964

Table 6
Annual averages of aerosol lifetime, burden, optical depth and direct radiative forcing

Aerosol constituent	Lifetime (day)	Burden (mg m^{-2})	Optical depth	Direct radiative forcing (Wm^{-2})
Sulphate ^a	3.60 ± 2.30	4.02 ± 3.98	0.026 ± 0.026	-1.11 ± 1.06
Organic matter	4.22 ± 3.92	4.28 ± 5.81	0.025 ± 0.034	-0.58 ± 0.87
Black carbon	5.49 ± 4.39	1.01 ± 1.24	0.010 ± 0.012	0.26 ± 0.32
Fly-ash	6.11 ± 10.33	7.23 ± 14.61	0.012 ± 0.023	-0.25 ± 0.59
Mineral	6.11 ± 10.33	1.91 ± 4.13	0.003 ± 0.008	-0.06 ± 0.20
Total		18.4 ± 22.1	0.08 ± 0.06	-1.73 ± 1.93

^aVenkataraman et al. (1999).

the estimated aerosol lifetime, burdens, optical depth and direct radiative forcing (Table 6). The uncertainties in all input parameters are propagated into an estimated uncertainty in the output variables for each aerosol constituent and the total $\text{PM}_{2.5}$ aerosol (120% for aerosol burden, 80% for optical depth and 115% for direct radiative forcing). The variance in the direct radiative forcing from the total aerosol is explained by the emission terms of each constituent, in the proportion of its burden. The variance in the direct radiative forcing for each constituent is explained largely by the rates of emission and wet deposition (in terms of rainfall and scavenging ratio). Dry deposition was assumed to occur based on a single deposition velocity for each aerosol constituent. Uncertainties in regional cloud cover, surface wind velocities and assumed optical parameters and albedo do not significantly affect the estimated direct radiative forcing. Therefore, improvement is required in the emissions estimates and scavenging ratios, to increase confidence in these predictions.

3.2. Aerosol lifetime

The lifetime of aerosols depends on transport and removal mechanisms (advection, dry deposition and wet deposition). Calculated lifetimes of black carbon and organic matter are low in May to October with efficient wet deposition, and high in November to April (Figs. 2a and b). The annual average of monthly mean lifetimes is 5.5 ± 4.4 day for black carbon and 4.2 ± 3.9 day for organic matter. These are in good agreement with the globally averaged lifetimes of 3.9–7.8 and 3.9–4.5 day, respectively for black carbon and organic matter (Cooke and Wilson, 1996; Liousse et al., 1996; Cooke et al., 1999). Organic matter lifetimes are lower than black carbon, because of its hydrophilic nature (higher scavenging ratios) and consequent greater wet deposition. Organic matter lifetimes are comparable to previously estimated sulphate lifetimes, with wet deposition being the predominant removal mechanism (Venkataraman et al., 1999).

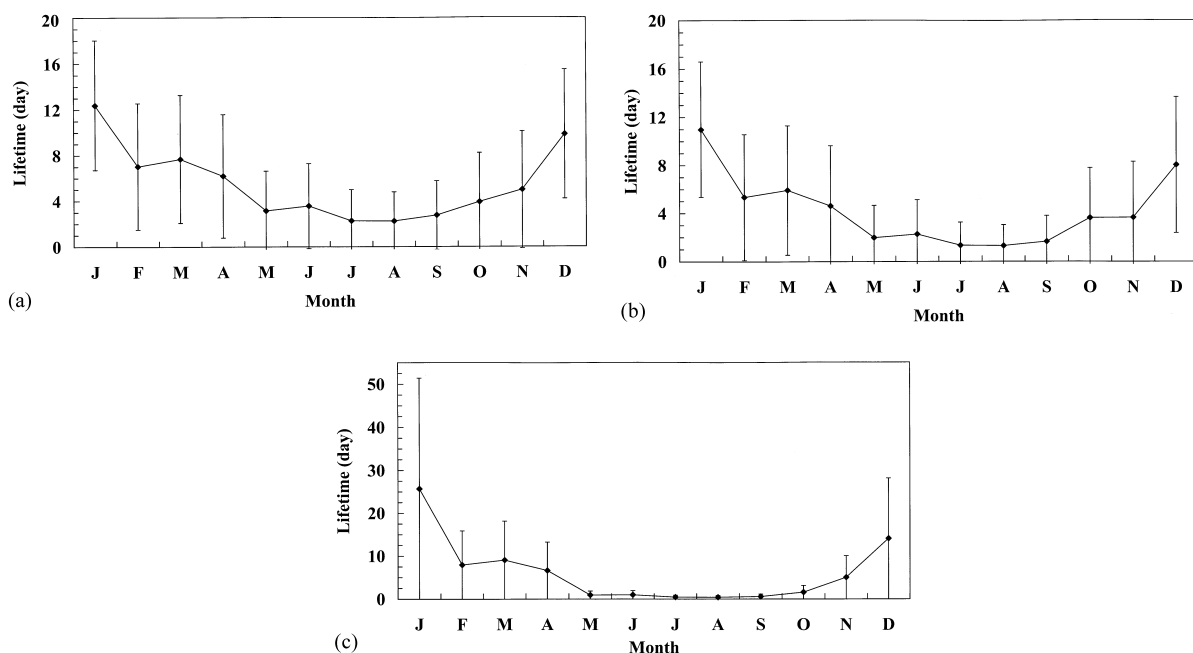


Fig. 2. (a) Black carbon aerosol lifetimes range 3–12 day. Removal is not efficient either by dry deposition (small particle size and low deposition velocity) or by wet deposition (hydrophobic and low scavenging ratios of 18–350). (b) Organic matter aerosol lifetimes range 2–11 day. Wet deposition is more efficient than for black carbon because of hydrophilic nature (scavenging ratios 35–760). (c) Fly-ash and mineral aerosol lifetimes range 0.4–26 day. Fly-ash and clay-mineral aerosols were assumed to have the same dry deposition (0.018 cm s^{-1}) and wet deposition (scavenging ratios 500–1000).

The fly-ash and mineral aerosols ($0\text{--}2.5 \mu\text{m}$ diameter) lifetimes are governed by wet deposition, because of their low dry deposition velocity (0.018 cm s^{-1}). The resulting lifetimes range from 0.4 to 26.0 day (Fig. 2c) with an annual average of 6.1 ± 10.3 day, in reasonable agreement with the global average lifetime (13 day), calculated from the detailed dust transport models (Tegen and Fung, 1995; Tegen et al., 1997).

3.3. Aerosol burden

The integrated column burden of aerosols is proportional to the lifetime and emission strength. Total monthly mean aerosol burdens are high, $14.2\text{--}54.4 \text{ mg m}^{-2}$ during November to April, and low, $4.9\text{--}10.0 \text{ mg m}^{-2}$ during May to October (Fig. 3). Annual average of total aerosol burden is $18.4 \pm 22.1 \text{ mg m}^{-2}$. The highest relative contribution to the total aerosol burden is from fly-ash (40%), followed by organic matter (23%) and sulphate (22%) (Fig. 4). While fly-ash emissions contribute only 23% of total aerosol emissions, its longer lifetime results in a 40% contribution to the total burden. Mineral aerosol emissions are higher (25% of total emissions), but their contribution to total burden is only 10%. The box model in its present form would underestimate mineral aerosol burdens, because of neglecting

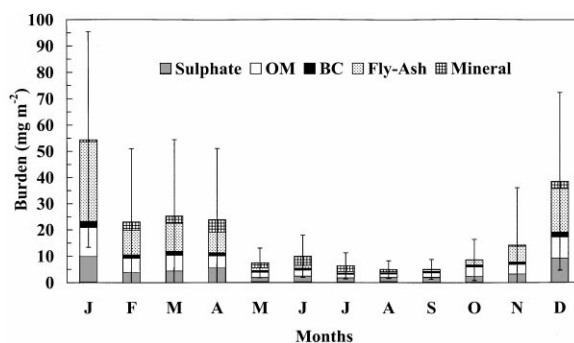


Fig. 3. Monthly mean aerosol burdens range ($5\text{--}54 \text{ mg m}^{-2}$) with lows in monsoon months and highs in the dry season.

the spatial distribution of aerosol emissions and rainfall. Mineral aerosol fluxes are higher in desert regions in north India, where the monsoon does not set in until late-July. Our model imposes spatially averaged rainfall over the entire Indian region, which was high in 1990 from May to August during the monsoon. A large part of the mineral aerosol emissions (80%) occur during this period, thus over-estimating wet deposition and under-estimating burden.

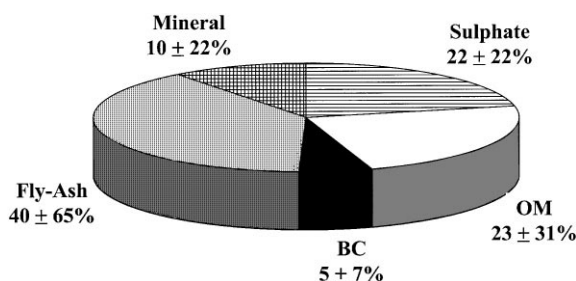


Fig. 4. Relative contribution to annual average aerosol burden ($18.4 \pm 22.1 \text{ mg m}^{-2}$) is highest from fly-ash followed by organic matter and sulphate. The mineral aerosol burden is underestimated in this model because of neglecting the spatial distribution of emissions and rainfall.

3.4. Aerosol optical depth

Optical depth of aerosol constituents, calculated as the product of the burden and the mass extinction coefficient (Table 5), assume that aerosol species do not interact with each other. The total monthly mean aerosol optical depths are low during May–October (0.04–0.05) and high during November–April (0.07–0.19), with an annual average of 0.08 ± 0.06 (Fig. 5).

These compare well with the measured optical depths of 0.07–0.17 (at $0.55 \mu\text{m}$) at Kaashidhoo (4.965°N , 73.466°E , which falls just outside the southwest corner of the box-model area of $8\text{--}36^\circ\text{N}$ and $70\text{--}92^\circ\text{E}$), during the February and March, 1998 (Satheesh et al., 1999). The Indian subcontinent was identified as the major source of aerosols from long-range transport. The estimated optical depth is in the lower part of the range of the measured total aerosol optical depth at five stations in India (0.10–0.80 during 1986–1989) (Krishna Moorthy et al., 1993). Our estimate does not include the contribution from natural mineral aerosols, sea salt, water vapour, other secondary aerosols (e.g. nitrate, ammonium) and gases to total optical depth and would therefore be lower than the measurements. Our model is unable to replicate the seasonal variation of a measured summer maximum, which has been attributed to the increased mechanical production of natural aerosols, and a winter low to aerosol removal from NE-monsoon rains. The seasonal variation is not replicated because our model neglects the spatial distribution of aerosol emissions and rainfall and imposes a spatial average rainfall over the entire Indian region. A large part of the mineral aerosol emissions occur during May–August, and the high monsoon rainfall over-estimates wet deposition and under-estimates the resulting burdens and atmospheric effects. A spatially resolved model would estimate these effects more accurately.

The highest relative contribution to annual average aerosol optical depth is from sulphate (36%) followed by organic matter (33%) and black carbon (14%) (Fig. 6),

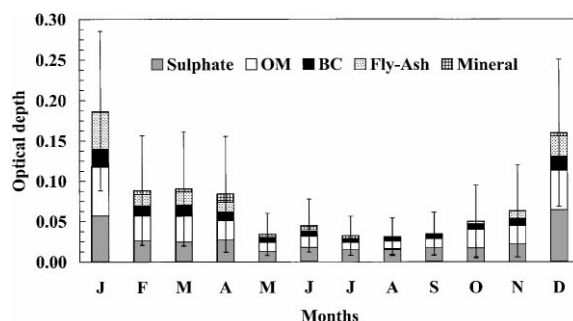


Fig. 5. Monthly mean aerosol optical depth ranges 0.04–0.19 with monsoon lows and dry season highs.

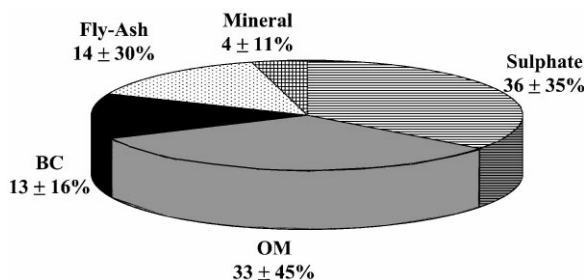


Fig. 6. Relative contribution to annual average aerosol optical depth (0.08 ± 0.06) is greatest from sulphate and organic matter followed by fly-ash and black carbon.

accounting for 82% of aerosol optical depth. This is attributed due to higher burdens and higher mass extinction coefficients of these aerosols as compared to the mineral aerosols. Though, mineral aerosols contribute 25% of the total emissions, their contribution to optical depth is only 4%, because of the underestimated burdens.

We compare our estimates of relative contribution to optical depth for February and March to that from an aerosol model for the tropical Indian Ocean (Satheesh et al., 1999). Using average aerosol mass concentrations at Kaashidoo for sea salt, nitrate, nss-sulphate and ammonium, dust and ash, measured optical depth and radiation flux, Satheesh et al. (1999) estimated the relative contribution of these species and black carbon to visible aerosol optical depth, and assigned the balance to “missing” or unaccounted organic matter. Their estimated contributions were organic matter (24%), nss-sulphate + ammonium (35%), black carbon (13%), ash (10%) and dust/mineral aerosol (18%). Our estimates for these species, organic matter (35%), anthropogenic sulphate (28%), fly-ash (17%) and black carbon (15%) are in reasonable agreement with the previous study. Our estimate of dust/mineral aerosol contribution to optical depth (5%) is much lower than that of Satheesh et al. (1999) (18%). This results from our under-estimate of

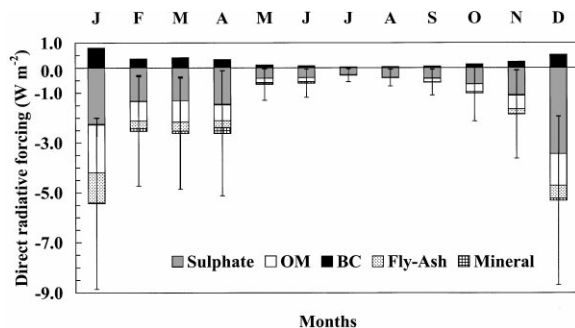


Fig. 7. Monthly mean direct radiative forcing ranges $+0.1$ to $+0.7 \text{ W m}^{-2}$ from black carbon and -0.8 to -4.8 W m^{-2} from other aerosol constituents.

mineral aerosol burdens, because of neglecting the spatial distribution of emissions and rainfall. Our estimate of organic matter contribution to optical depth is 35% and would be revised downward once the mineral aerosol estimate is corrected. This estimate, made from a source/sink mass balance, corroborates the likely large contribution of organic matter and fly-ash aerosols to atmospheric optical effects.

3.5. Direct radiative forcing

The estimated monthly mean direct radiative forcing is low during May–October (-0.27 to -0.86 W m^{-2}) and high during November–April (-1.61 to -4.64 W m^{-2}) with an annual average of $-1.73 \pm 1.93 \text{ W m}^{-2}$ (Fig. 7). The direct forcing from all aerosol constituents except black carbon aerosols is negative resulting in net cooling. Organic matter aerosol forcing ($-0.58 \pm 0.87 \text{ W m}^{-2}$) is negative and comparable to that from sulphate ($-1.10 \pm 1.00 \text{ W m}^{-2}$). The radiative forcing from the mineral aerosols is negligible ($-0.06 \pm 0.20 \text{ W m}^{-2}$) as compared to other aerosol constituents. Annual average forcing from black carbon is $+0.26 \pm 0.32 \text{ W m}^{-2}$ resulting in net heating. Sulphate aerosols contribute 49% of the total forcing, followed by organic matter (26%) and fly-ash (11%) (Fig. 8). Once again, the importance of organic matter and fly-ash aerosols to atmospheric effects is indicated.

4. Conclusions

A box model has been used to compare the estimates of burden, optical depth and direct radiative forcing from anthropogenic $\text{PM}_{2.5}$ aerosol constituents over the Indian subcontinent. An aggregate estimate of $\text{PM}_{2.5}$ emissions was made, for the first time, for this region. The total anthropogenic combustion derived aerosol

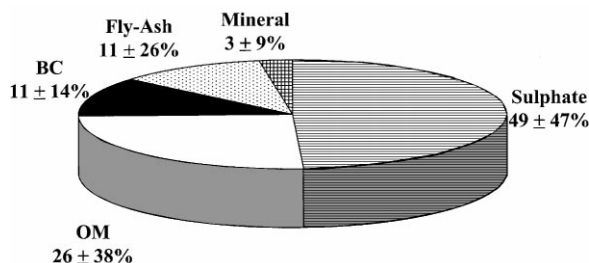


Fig. 8. Relative contribution to annual average direct radiative forcing ($1.73 \pm 1.93 \text{ W m}^{-2}$) is highest from sulphate. Significant scattering from organic matter and fly-ash aerosols has been estimated. Absorption by black carbon results in a positive forcing of $0.26 \pm 0.32 \text{ W m}^{-2}$.

emissions ($\text{PM}_{2.5}$) from India for 1990 are 9.4 Tg yr^{-1} , with fossil fuel combustion and biomass burning accounting for 62 and 38%, respectively. Adding the anthropogenic mineral aerosols from land disturbance ($0\text{--}2.5 \mu\text{m}$ diameter) to the above results in total aerosol emissions of 12.6 Tg yr^{-1} . The contribution from various aerosol constituents is 28% sulphate, 20% organic matter and 4% back carbon. Coarse fraction aerosols were not included, as they would not contribute to regional aerosol burdens and atmospheric effects.

The monthly mean aerosol burden ranges from 14.2 to 54.4 mg m^{-2} with an annual average of $18.4 \pm 22.1 \text{ mg m}^{-2}$, with the largest contribution from fly-ash from high ash-content coal burning (40%), followed by organic matter (23%) and sulphate (22%). The alkaline constituents of fly-ash could neutralise rainfall acidity and contribute to the observed high rainfall alkalinity in this region (Parashar et al., 1996).

Aerosol optical depths vary from 0.04 to 0.19, with an annual average of 0.08 ± 0.06 , sulphate accounting for 36%, organic matter for 33% and fly-ash for 14%, in agreement with the source contribution estimates of Satheesh et al. (1999). The mineral aerosol contribution (5%) is much lower than the previous estimate (18%) because of wet deposition from high rainfall in the months of high emissions and the complete mixing assumption in the box model. Our model results corroborate the potentially large contributions of organic matter and fly-ash aerosols to atmospheric optical effects in the Indian region.

The annual average radiative forcing is $-1.73 \pm 1.93 \text{ W m}^{-2}$. The major contributors are sulphate aerosols (49%) followed by organic matter (26%), black carbon (11%) and fly-ash (11%). Once again, the importance of organic matter and fly-ash to atmospheric radiative effects is indicated.

The uncertainties in estimated parameters (aerosol lifetime, burden, optical depth and direct radiative forcing) resulting from uncertainties in input parameters (emission

rates, deposition and optical parameters) range 80–120%. The uncertainties in emission and wet deposition rates contribute significantly to this variance. Therefore, improvement is required in the emissions estimates and scavenging ratios, to increase confidence in these predictions.

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